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Laser Ignition of Solid Propellants: I. Ignition Delays

Arthur Cohen
Richard A. Beyer

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13. ABSTRACT (Maximum 200 words) Ignition experiments on nitrate-ester and nitramine propellants have been conducted in a closed bomb. The ignition source was a variable (<100 W) cw CO ₂ laser. Ambient conditions were room temperature and atmospheric pressure. Air, argon, and nitrogen gases were used. Times to detect emission and changes in emission rate which are attributed to onset of flamespreading have been compared with ignition delay calculations using a condensed phase radiant ignition model. Reasonable values for thermochemical properties and experimental determined values for chemical kinetic parameters and optical properties were used for the calculations. The flux dependence of the initial emission delays were in reasonable agreement with model predictions for nitrate-esters but not for nitramines. Gas phase (composition) effects on initial emission were observed with XM39, an RDX nitramine, but not with nitrate-esters. Flamespreading delays showed gas composition effects with all propellants. These results suggest that (for all propellants) the effects of gas phase reactions on flamespreading and (for nitramines) on initiation need to be included (explicitly) in radiant ignition models for predicting pressurization in gun chambers.				
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1. INTRODUCTION

One of the objectives of the LIGHT program is to determine the feasibility of replacing gun igniter systems with lasers. This is expected to increase safety and survivability and to improve the performance of gun systems. Models of the laser-solid propellant ignition process which can be used to calculate the initial gun chamber pressurization will help achieve this objective. By comparing model calculations with results from closed bomb experiments, the ability of (thermal) radiative ignition models to predict the response of solid propellants to laser irradiation can be evaluated.

Many radiative ignition models are available for evaluation (Kulkarni, Kumar, and Kuo 1982; Hermance 1984). They are usually classified according to the phase(s) in which the self-accelerating exothermic reaction(s) associated with the ignition process occurs (i.e., condensed, gas, heterogeneous).

Most models are one dimensional and have ignition criteria based on transient temperature behavior. The physical and chemical processes which describe the "ignition" mechanism differ even in models of the same class (Kulkarni, Kumar, and Kuo 1982). To compare predictions with experimental data usually requires numerical solutions of the equations used to describe the details of these processes. Analytic solutions are possible through use of simplifying assumptions (Williams 1966). Phenomonological ignition models do not attempt detailed descriptions of the ignition processes, and the corresponding equations, in some cases (Vilyunov and Zarko 1989; Anderson 1972), have analytic solutions.

It is generally difficult to evaluate radiative ignition models due in part to difficulties of matching the experimental and model boundary conditions and relating diagnostics and model ignition criteria. In addition, the physical/optical properties and heat release rates which are required as inputs to the model are not well known. Laser ignition models may have an additional problem (i.e., describing the details of the laser energy [thermal and nonthermal] interactions with the propellant and with reaction products).

The initial results of an investigation of laser ignition of some nitrate ester (M9, M30, JA2) and nitramine composite (XM39, HMX2) solid propellants under ambient conditions will be presented, and correlations of emission and flamespreading delay measurements with predictions of a phenomonological radiant ignition model will be discussed.

2. IGNITION MODEL

The model (referred to as "approximate") is described in Vilyunov and Zarko (1989). It considers condensed phase (surface) reactions and in-depth energy absorption but neglects energy losses (i.e., those losses due to surface [film] heat transfer and emission), propellant consumption, and does not consider the effect of laser interruption (deradiation). The model is one dimensional (semi-infinite solid). It assumes that flux is continuous and uniform and that energy production can be described by a first-order (global) surface reaction. The relevant energy equation and boundary conditions used for (transparent) solids are:

$$c \frac{\partial T}{\partial t} = \alpha c \frac{\partial^2 T}{\partial x^2} - \frac{1}{\rho} \frac{\partial q}{\partial x} + Qz \exp(-E/RT)$$

$$T(x, 0) = T_0; \quad \frac{\partial T}{\partial x}(0, t) = 0; \quad T(\infty, t) = T_0,$$

where $q = q_0 \exp(-nx)$, q_0 = transmitted surface flux, n = absorption coefficient, T = temperature, t = time, x = distance from irradiated surface, T_0 = ambient temperature, α = thermal diffusivity, ρ = density, and c = specific heat. The last term in the energy equation is the Arrhenius global heat release rate during ignition (E = activation energy, z = first-order pre-exponential factor, and Q = heat of reaction).

The ignition delay (t_{ig}) is the sum of (inert) "heat up" (t_h) and (chemical) induction (t_{ch}) times. The surface temperature (T_h) and t_h are determined by finding the temperature at which the time derivatives obtained from analytic solution of the energy equations corresponding to inert (radiant) heating ($Q = 0$) and to adiabatic (spatially independent) reaction ($\alpha = q_0 = 0$) become equal. T_h represents the chemical reaction temperature at which the (specific) rate of chemical energy production equals the radiant energy absorption (at the surface). t_h is the time required to reach this temperature. At later times, due to the Arrhenius exponential temperature dependence, chemical rates dominate and it is assumed that surface reaction occurs under adiabatic conditions. The chemical induction time is obtained from the adiabatic solution and is defined (in this model) as the time (with respect to t_h) for time derivative to become infinite. Approximations to these analytic solutions (given in Vilyunov and Zarko 1989) were used to calculate T_h and the corresponding t_h and t_{ig} as a function of radiant flux. The inert approximation (for T_s and t_h) is valid under conditions when characteristic lengths for heat conduction during heat up are much greater than those for absorption ($n\sqrt{\alpha t_h} \gg 1$). The adiabatic approximation (for t_{ch}) is valid

when activation energies are much greater than thermal energies ($E/RT_h \gg 1$). For convenience, the derivation (along with the energy equations and their solutions) of these approximations is shown in the Appendix. In all calculations, $T_0 = 298$ K, and for all propellants, the following values were used for the thermophysical properties: $\alpha = 0.001$ cm²/s, $c = 0.35$ cal/g-K, and $\rho = 1.6$ g/cm³. A distinguishing feature of the model is the prediction of a (chemical kinetic-dependent) flux level above which $t_{ch} \ll t_h$ (i.e., $t_h = t_{ig}$). This "approximate" theory has been previously used to predict the dependence of "ignition" delays for secondary explosives (HMX, RDX) on laser flux (Vilyunov and Zarko 1989). Reasonable success was obtained using experimentally determined values for the input parameters.

3. EXPERIMENTAL

A cw CO₂ (10.6 μ m) laser (Synrad model No. 51) with variable power (nominal power = 10–100 W) was used as the ignition source. Power was measured with a thermopile (Coherent Radiation model 201). The estimated measurement error is $\pm 10\%$. The responses of several propellants at different power (flux) levels and in different gaseous environments have been briefly examined. Data have been obtained for the following propellants: M9 (homogeneous NC double base), JA2 (homogeneous NC triple base), M30 (composite NC triple base), XM39 (RDX composite), and HMX2 (experimental HMX composite). The samples were small cylinders approximately 3/16 in long and 0.25 in diameter. The M30 and XM39 samples were cut from grains (perforated with graphite surfaces), and the others were cut from solid strands. The beam (diameter about 3 mm) was directed horizontally along the propellant axis and covered approximately 25% of the sample (plane) surface. Pulse lengths were varied from 0.1–15 s. Delays to emission and flamespreading, denoted by t_e and t_f , were measured from emission traces recorded by a photomultiplier (300–800 nm response), which viewed the region near the irradiated surface. t_e represents the time (from start of irradiation) to detect emission, and t_f , which is a more subjective measurement, represents the time to observe a change in the slope of the emission signal consistent with onset of a relatively steady state process. This change is assumed to indicate the start of the flamespreading process. Estimated measurement errors for t_e and t_f are $\pm 5\%$ and 15% , respectively. Most measurements were made in a closed bomb (inside dimensions 3 in diameter by 9 in tall). The location and sensitivity of the photomultiplier were not changed for these experiments. The bomb has optical access from four sides, with one containing a ZnSe window for transmitting the 10.6- μ m radiation. The bomb was initially filled with air, nitrogen, or argon. Ambient conditions were room temperature and atmospheric pressure.

An infrared (IR) microscope-spectrometer (Spectra-Tech Plan 40/Matson Polaris) was used to measure propellant reflectivities and transmittivities at 10.6 μ m. Propellant reaction products were analyzed by gas chromatography and FTIR spectrometry.

4. EXPERIMENTAL RESULTS

4.1 Optical Properties. Bouguer's law was used to derive the absorption coefficients from the dependence of transmittivity on propellant width. This implicitly assumes scattering coefficients \ll absorption coefficients. At present, the absorption coefficients for M9, JA2, and XM39 have been determined and are 536, 250, and 756 cm^{-1} , respectively. In the visible region, the transparency of JA2 is much greater than M30 and HMX2. If this is also true at 10.6 μm , then the value of $n = 250 \text{ cm}^{-1}$, which has been used for the absorption coefficient of M30 and HMX2 in calculations, will represent a lower bound. This leads to an upper bound for the calculated ignition delays.

The results of the initial reflectivity measurements indicate that at 10.6 μm all propellants reflect less than 10%. For comparison with calculations the measured flux values were corrected using a reflectivity value of 0.08.

4.2 Gas Product Analysis. At present, only a cursory examination of the results from gas analyses has been made. The major difference between samples from flamespreading and non-flamespreading experiments, besides concentration levels, appears to be the higher CO/CO_2 ratio for the latter.

4.3 Laser Ignition. The following figures contain photomultiplier records selected to show some of the complexities occurring during laser ignition experiments, and graphs which compare the emission and "flamespreading" delays obtained from such records with predictions of the "approximate" ignition model. For some experiments, "flamespreading" delays (t_f) were greater than the laser pulse duration (t_l). In such cases, t_l , which is a minimum lower bound for t_f , was used for the comparisons. In some cases, data represent average values of several measurements.

Listed in Table 1 are the laser power (P), t_l , and the corresponding delays (t_e , t_f , t_d) obtained from the photomultiplier signals shown in Figures 1, 3, 5, and 7. t_d is an estimate of the final emission decay time.

4.3.1 Nitrocellulose Propellants. Figure 1 shows the effect of flux levels on emission during ignition and flamespreading of M9 in nitrogen at ambient conditions. Time is relative to start of the laser irradiation. The points e, f, and d locate the emission delay, t_e ; "flamespreading" delay, t_f (corresponding to a slope change in the emission-time signal); and final emission decay time, t_d . Both t_e and t_f (conceivably related to t_h and t_{ig} , respectively) decrease as power increases. The M9 emission decay at d1 and d2 is due primarily to consumption of propellant.

Table 1. Laser Operating Conditions and Delay Measurements for Figures 1, 3, 5, and 7

Figure	Signal	Laser Power (W)	t_l (s)	t_e (s)	t_f (s)	t_d (s)
1	1	2.5	5.80	0.19	0.30	1.65
	2	8.6	1.10	0.03	0.09	1.40
4	1	1.7	4.34	1.93	4.85	4.30
	2	8.6	1.67	0.05	(0.12)	1.70
6	—	4.2	2.72	0.04	1.37	3.60
8	1	8.6	0.62	0.18	2.10	0.65
	2	8.6	0.20	0.11	—	0.22

t_l = laser pulse duration

t_e = emission delay

t_f = flamespreading delays

t_d = estimate of final emission decay time

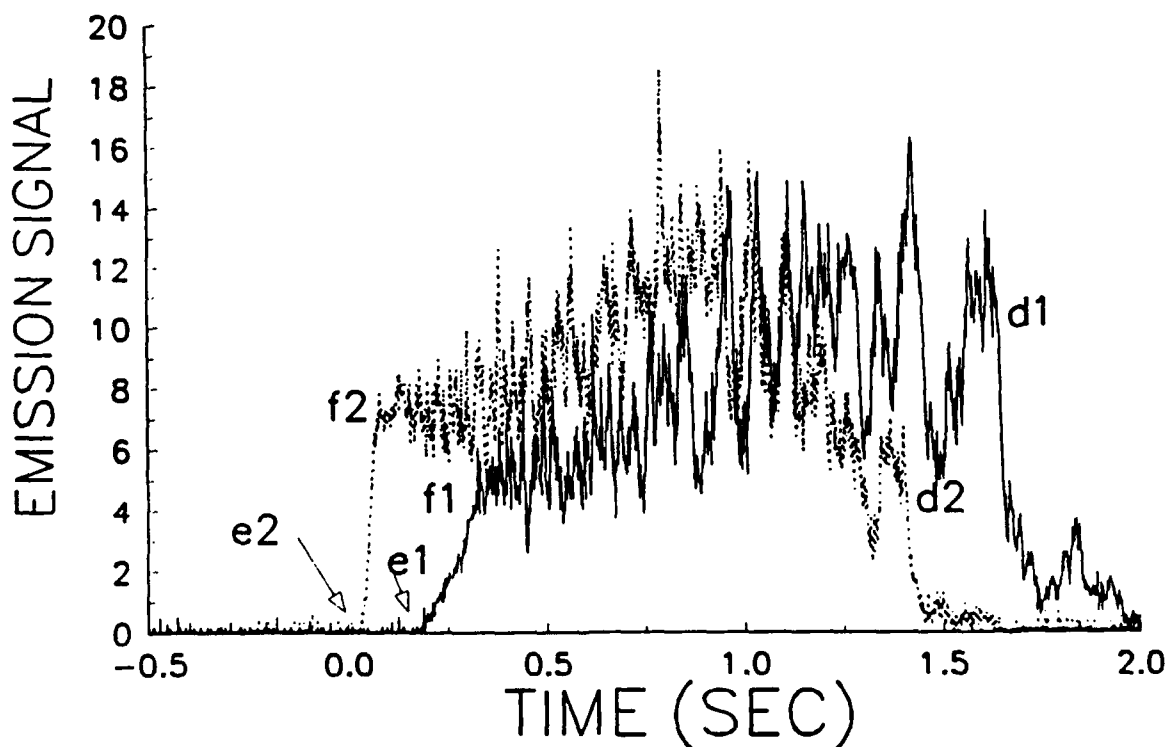


Figure 1. Photomultiplier records showing effect of flux level on emission from M9 in N_2 .

The lines in Figure 2 are the ignition delays calculated from the model as a function of flux level for different heat release rates. "N" refers to a rate (given in Vilyunov and Zarko 1989) determined from radiant (black body) ignition experiments with the Russian double-base propellant N (0.58 NC, 0.28 NG, and 0.12 DNT), and "M9" refers to a rate determined from shock tube experiments with M9 (0.58 NC, 0.40 NG) powders in nitrogen ($T = 600\text{--}700\text{ K}$ and pressure = $0.9\text{--}3.5\text{ atm}$) (Cohen and Holmes 1982). For N and M9, $Q_z\text{ (cal/g-s)} = 2.5\text{E}16$ and $2.6\text{E}11$, respectively. The corresponding values for $E\text{ (kcal/mole)}$ are 35 and 22. The rate for N propellant was determined from measurements of delays to observe a spike in the surface (thermocouple) temperature. The spike signaled the start of a rapid temperature increase presumably due to flamespreading. The rate for M9 was determined from emission delay measurements using small ($<10\text{ }\mu\text{m}$) particles uniformly heated (primarily) by conduction from reflected shock gas.

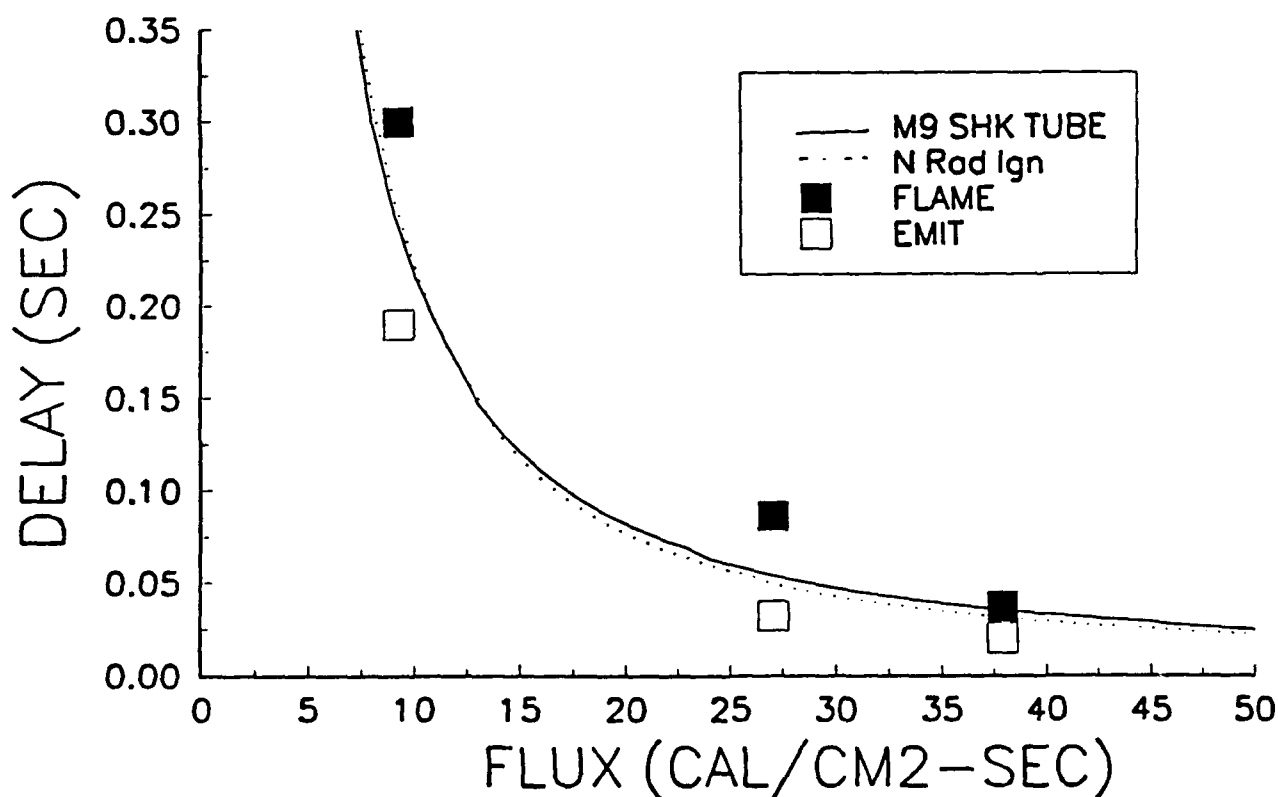


Figure 2. Comparison of emission and flamespreading delays for M9 in N_2 with calculations using heat release rates determined from radiative ignition and shock tube experiments.

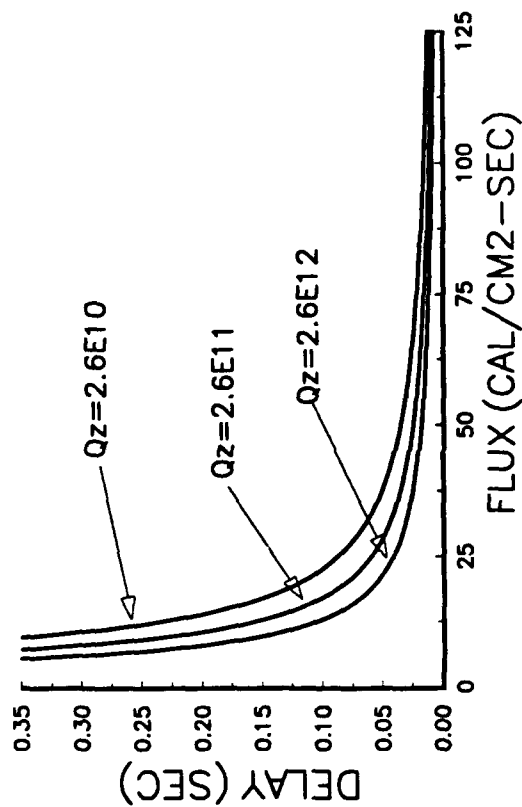
At flux levels $>10 \text{ cal/cm}^2\text{-s}$, (i.e., for calculated delays $<0.3 \text{ s}$), differences between the calculated ignition delays are less than estimated measurement errors (flux $\pm 10\%$, $t_e \pm 5\%$, and $t_f \pm 15\%$).

The data are the t_e and t_f measurements with N_2 (in the bomb) initially at ambient conditions. Considering measurement errors and the uncertainties in thermophysical properties, both t_e and t_f appear to be in reasonable agreement with predictions of the "approximate" model.

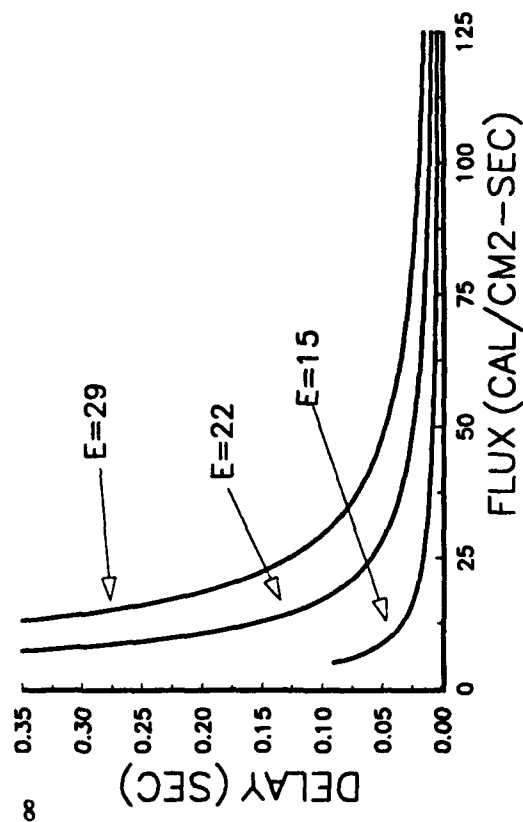
Figures 3a, 3b, and 3c show the flux dependence of the t_{ig} calculation on the kinetic parameters Q_z and E and the absorption coefficient n . The middle lines correspond to the one calculated for M9 in Figure 2a ($Q_z = 2.6\text{E}11$, $E = 22$, and $n = 536$). The dependence on kinetic parameters decreases markedly at higher flux levels ($>25 \text{ cal/cm}^2\text{-s}$). The dependence on n decreases markedly for absorption lengths $(1/n) < 20 \mu$.

Figure 4 (and Table 1) show the effects of flux level and pulse duration on JA2 response to deradiation. The photomultiplier signals are from experiments in Ar at ambient conditions. The values of t_e and t_f decrease with increasing power, similar to the M9 results, but at higher power (and higher energy) and shorter pulse length, the final emission decay at d2 occurs at laser shutoff, t_l (i.e., deradiation leads to extinguishment). The only evidence of reaction in the recovered sample is a crater formed (primarily) in the irradiated region. Emission, in this case, is due to local gasification (laser-assisted combustion) reaction. At lower power and longer pulse lengths, reaction decelerates at t_l (corresponding to d1 in this figure), but recovers. Self-acceleration leads to the sharp increase in emission at f1 which is assumed to be the start of flamespreading. The subsequent decay (not shown) in emission is the result of propellant consumption.

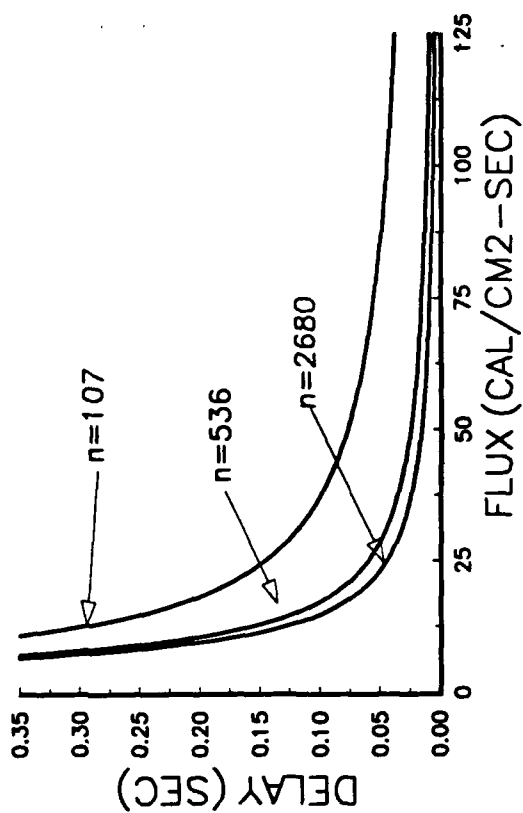
The data in Figure 5 are the emission and "flamespreading" delays for JA2 in air and Ar at ambient conditions. The t_l values were used in place of the t_f values for air at 40 and for Ar at $4 \text{ cal/cm}^2\text{-s}$. These t_l values are approximately 10% less than the corresponding t_f measurements. The data at $90 \text{ cal/cm}^2\text{-s}$ are from an experiment in room air. All others are from experiments in the bomb. The line is the calculated M9 ignition delay shown previously in Figure 2. The data near $40 \text{ cal/cm}^2\text{-s}$ suggest that substitution of air for argon shortens t_f but does not affect t_e . The differences between calculation and measurements are probably within estimated measurement/thermophysical properties errors for t_e at the higher flux levels, but the differences are much greater for t_f .



a. Kinetic pre-exponential Q_z (cal/g-s).



b. Kinetic activation energy E (kcal/mole).



c. Absorption coefficient n (cm^{-1}).

Figure 3. Effect of input parameters on calculated ignition delays.

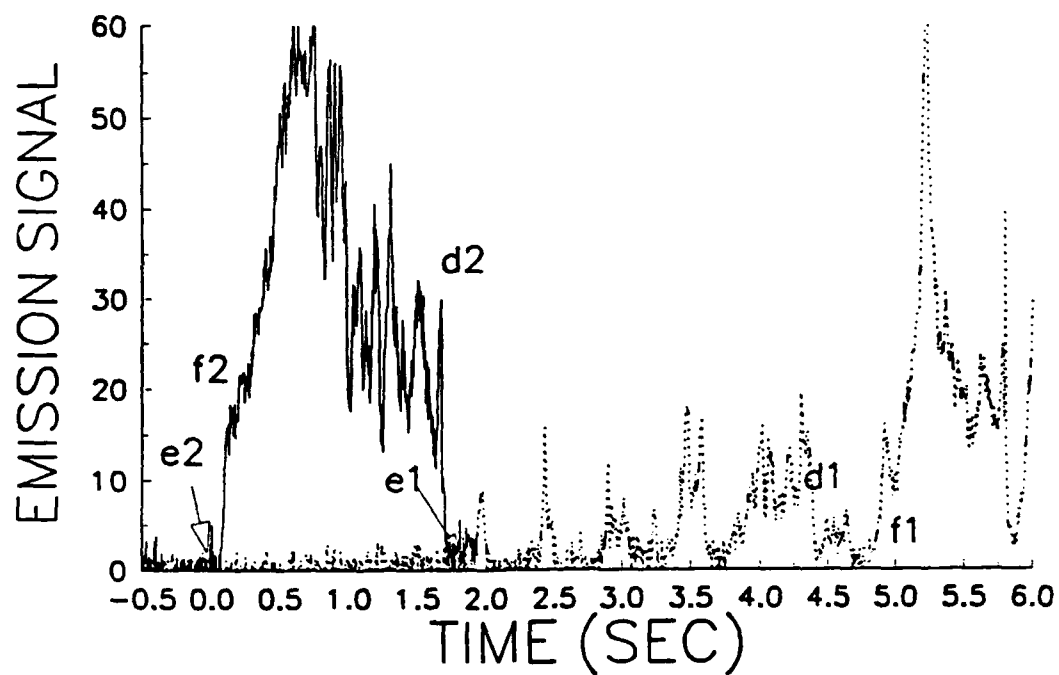


Figure 4. Photomultiplier records showing the effects of power and pulse length on reaction of JA2 in Ar.

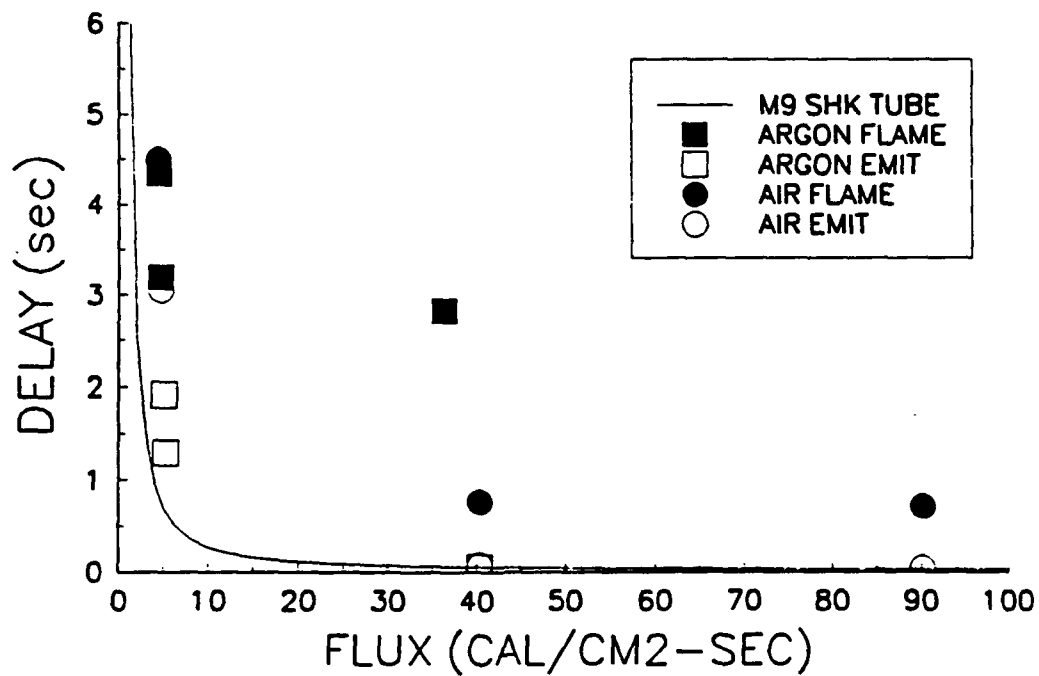


Figure 5. Comparison of emission and flamespreading delays for JA2 in Ar and air with calculations.

Figure 6 shows the emission signal during ignition of M30 in room air in which a transition from fizz (at f) to (visible) flame burning (at g) occurs. This transition has only been observed during irradiation (laser assisted) and differs from the transition to flamespreading with JA2, (at f1 in Figure 3) which occurs after deradiation. This suggests that absorption of laser radiation by M30 reaction products contributes to this transition.

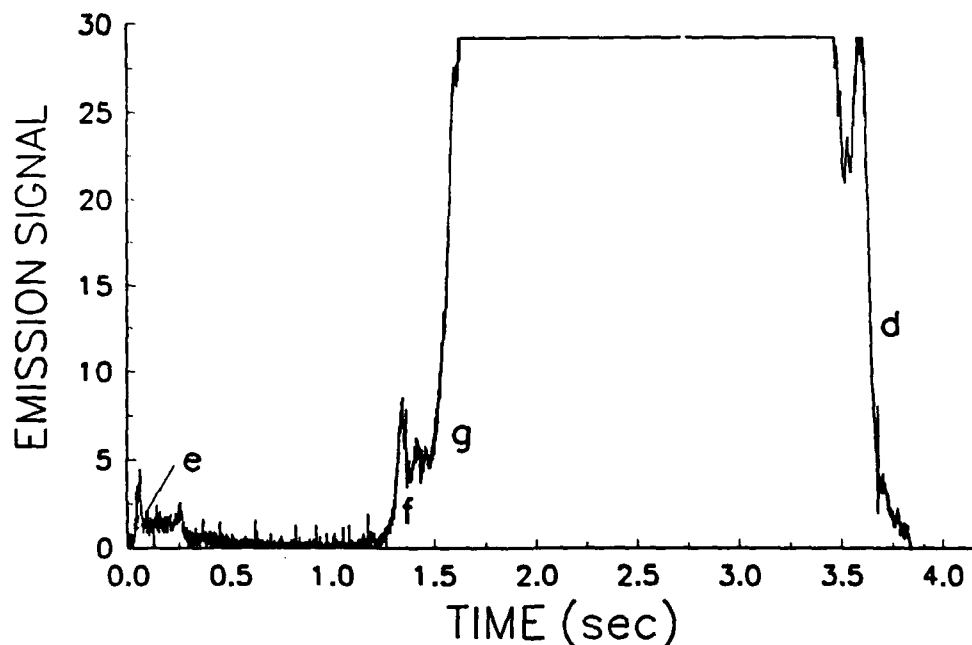


Figure 6. Photomultiplier records showing transition from fizz to flame burning for M30 in air.

The data in Figure 7 are the emission and "flamespreading" delays for M30 in room air and with N_2 in the bomb at ambient conditions. The line is the calculated M9 ignition delay. The results suggest that t_e values are not greatly different in air or N_2 , and, except at low flux values, are in reasonable agreement with the M9 calculations. The t_f values (near $35 \text{ cal/cm}^2\text{-s}$) are less in air than in nitrogen and are much greater than calculations.

4.3.2 Nitramine Propellants. Figure 8 (and Table 1) show the effect of pulse length on the response of XM39 to deradiation. The emission signals are from experiments at fixed power in air initially at ambient conditions in the bomb. The initial spike in the emission signals may be due to RDX reaction. The difference in t_e values may reflect the stochastic nature of the ignition process. The initial propellant

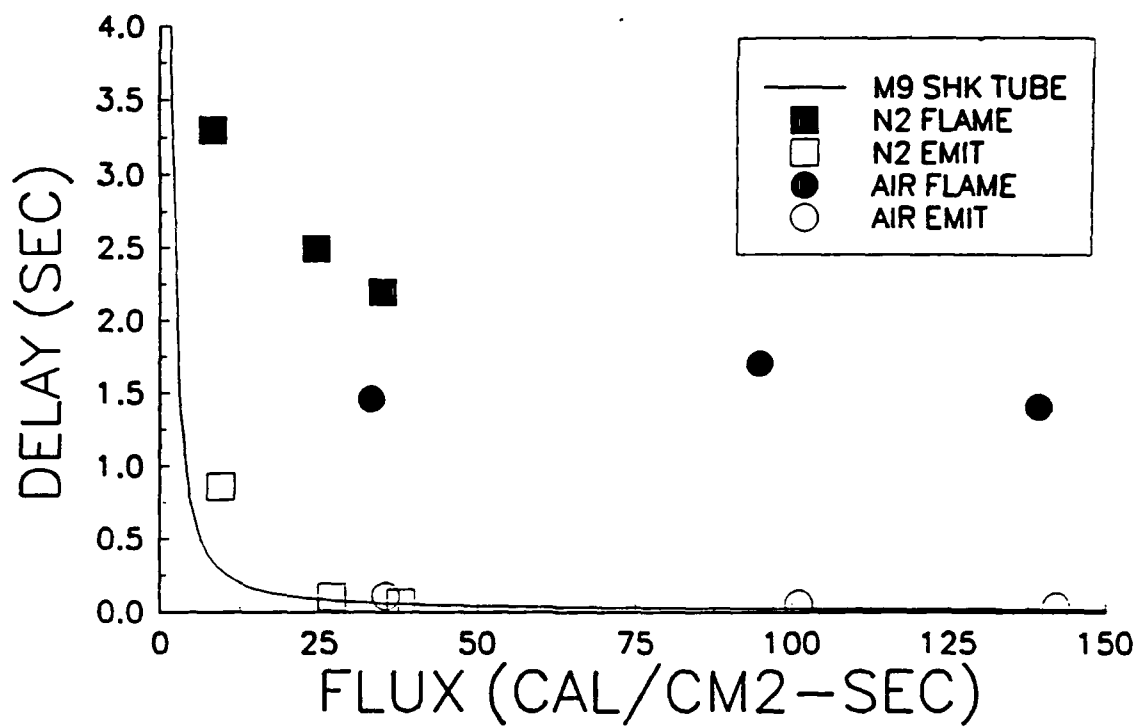


Figure 7. Comparison of emission and flamespreading delays for M30 in N₂ and air with calculations.

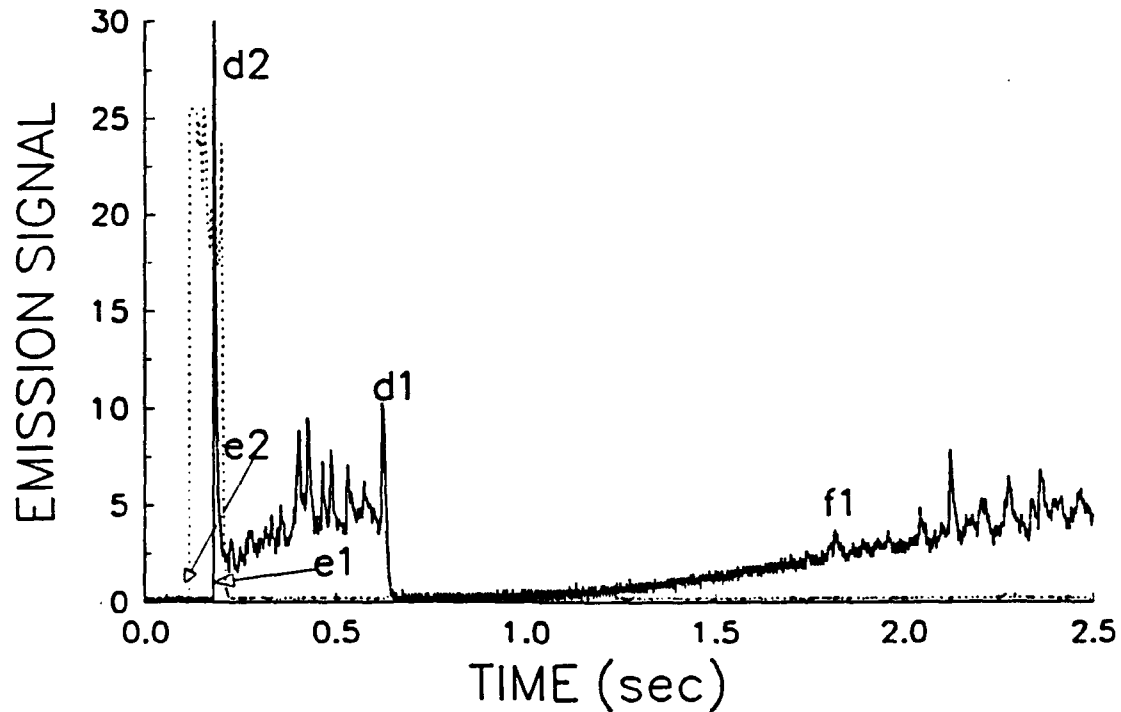


Figure 8. Photomultiplier records showing the effect of pulse length on reaction of XM39 in air.

response to deradiation is a sharp decay in emission. With the shorter pulse, reaction stops (at d2), leaving a crater at the irradiated surface. With the longer pulse, it recovers (at d1) and continues to self-accelerate. The final emission decay (not shown) is the result of propellant consumption.

The lines in Figure 9 are the calculated ignition delays as a function of flux levels using different heat release rates for XM39. "DSC" refers to a rate determined from DSC experiments with XM39 (Miller et al. 1988). In these experiments, the exothermic reaction was preceded by an endothermic reaction. For the calculations, the heat of the XM39 reaction was taken as the sum of both reactions and the heat release rate was taken as that of the exotherm. "Tcr" refers to a rate based on DSC experiments with RDX which was adjusted to give agreement between calculated and measured critical explosion temperatures for RDX (Rogers 1975). The rate for XM39 was obtained by multiplying the RDX rate by 0.75, which is approximately the mass fraction of RDX in XM39. The effect of the other constituents on the rate has been neglected for this calculation. The values of $Q_z(\text{cal/g-s})$ for "DSC" and "Tcr" are $5.46\text{E}16$ and $5.8\text{E}20$, respectively. The corresponding values for E (kcal/mole) are 38.2 and 47.1. For flux levels $>1 \text{ cal/cm}^2\text{-s}$, the differences between calculations are much less than measurement errors. The data are t_e values for XM39 with air and Ar, and t_i (pulse length) values (with air), all at ambient conditions in the bomb. Flamespreading was observed with air at flux levels $>25 \text{ cal/cm}^2$, but not with Ar (i.e., infinite t_i). The t_i values were approximately 60% of the corresponding t_e values. The t_e values for air are in reasonable agreement with calculations, except at low flux levels. The t_i values are probably greater than calculated ignition delays. Comparison of the results with Ar and air indicates that O_2 accelerates the XM39 reactions responsible for both emission and flamespreading. DSC experiments are with inert atmospheres. This suggests that the DSC kinetics are not appropriate for XM39 (or RDX) reaction in air. The observed agreement between t_e for air and calculations may be fortuitous. It appears that the model and/or rates do not represent laser-initiated reaction in XM39 (for flux levels $<47 \text{ cal/cm}^2\text{-s}$).

The lines in Figure 10 are the calculated ignition delays for HMX2 as a function of flux levels using different heat release rates. The line "SHK TUBE" refers to a rate for HMX2 determined from shock tube experiments with HMX powders in nitrogen ($T = 690\text{--}740 \text{ K}$ and approximate atmospheric pressure). "Tcr" refers to a rate based on DSC measurements with HMX, which have been adjusted to give agreement between calculated and measured critical explosion temperatures for HMX. The rates for HMX2 were obtained by multiplying the HMX rates by 0.8, which is the mass fraction of HMX in HMX2. The effect of the other constituents on the rate has been neglected for this calculation. The

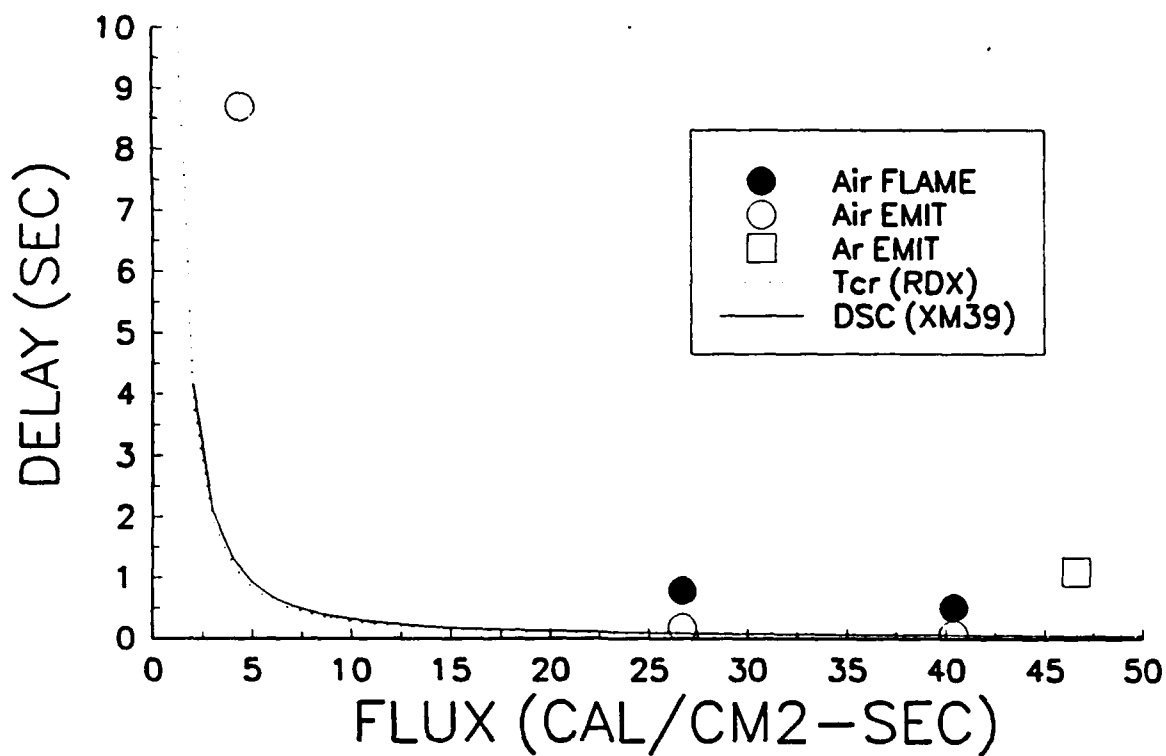


Figure 9. Comparison of emission and flamespreading delays for XM39 in Ar and air with calculations using heat release rates determined from DSC and critical temperature experiments.

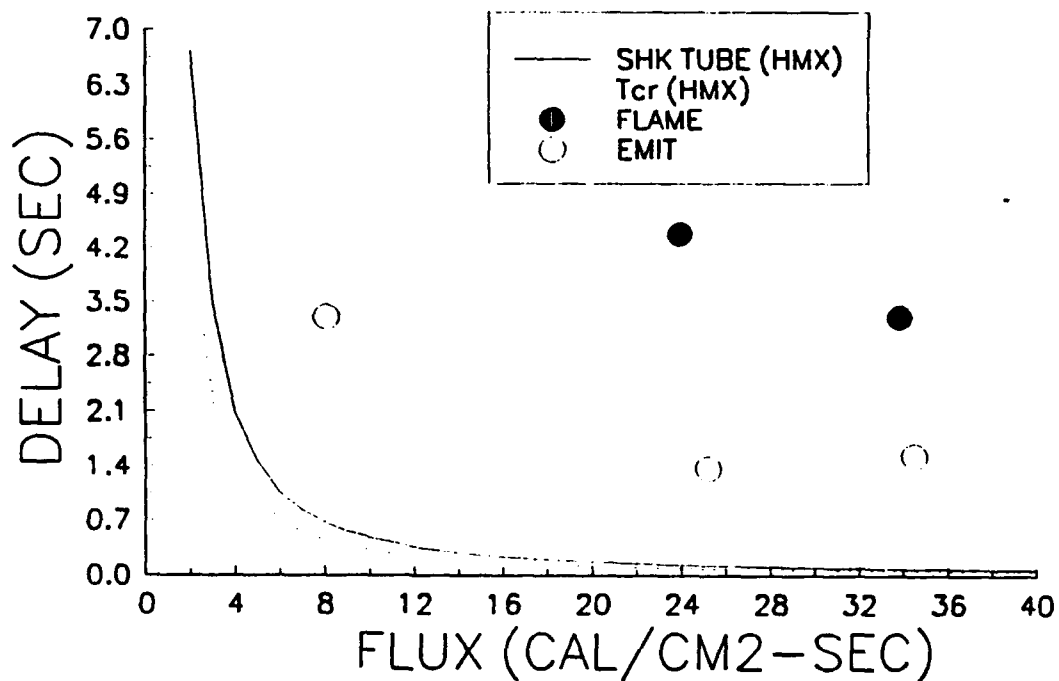


Figure 10. Comparison of emission and flamespreading delays for HMX2 in N₂ with calculations using heat release rates determined from shock tube and critical temperature experiments.

values of Q_z (cal/g-s) for "SHK TUBE" and "Tcr" are $1.5E17$ and $2.0E22$, respectively. The corresponding values for E (kcal/mole) are 44.3 and 52.7. Calculated delays are noticeably shorter for the DSC rate at flux levels <20 cal/cm²-s. The data are the emission and "flamespreading" delays for HMX2 in the bomb with nitrogen at ambient conditions. The values of t_e and t_f are much greater than the calculations, which indicates that the model and/or rates do not represent laser-initiated reaction in HMX2 (for flux levels <35 cal/cm²-s).

5. COMMENTS

At this stage of the investigation, there is insufficient data to draw many firm conclusions concerning the modifications of the "approximate" model for use in the LIGHT program. The following are comments concerning requirements of a "useful" laser ignition model and some conclusions derived from the data.

1. The comparison of ignition model predictions and experimental delays implicitly assumes that the delays represent times required for the propellant reactions to generate conditions which satisfy the ignition criterion. For the "approximate" model, this requires that chemical reaction rates become large (infinite). At present, the validity of this assumption for either the measured emission delays or "flamespreading" delays is not known.

2. Although boundary conditions of model (1-D, uniform flux distribution) and experiment (2-D Gaussian flux distribution) differ, the model (and input parameters) appears to be able to predict M9 response to the CO₂ laser irradiation. It appears that both t_e and t_f are in reasonable agreement with model predictions, although, in principle, the ignition delay is unique. Which (if any) of the experimental delays corresponds to the calculated ignition delay and reasons for this dual agreement are, at present, not known. It also appears that at high flux levels and when using kinetics determined for M9, the model can reasonably predict the emission delays, but not the "flamespreading" delays of other nitrate ester propellants. However, the model does not appear to be appropriate for nitramine composite propellants.

3. The decrease in the flux dependence of the calculated ignition delay (for all kinetic rates) as flux levels increase suggests that precise knowledge of initiation rates will not be critical inputs into models of flamespreading at high flux levels.

4. The substitution of air for inert gases leaves t_e values for nitrate ester propellants unchanged, but shortens t_f values. This suggests that reaction of gases (O_2) affect flamespreading but not initiation.

5. The substitution of Ar for air with XM39 (nitramine) propellant appears to increase t_e and t_f values. This suggests that the reaction of gases (O_2) affect both initiation and flamespreading.

6. The laser-assisted transition from fizz to flame burning with M30 in air (Figure 6) suggests the possibility that laser absorption by reaction products aids in the transition. Knowledge of absorption coefficients and concentrations of these products will be required to predict this behavior.

7. Observations of deradiation and ambient gas effects similar to present observations but at higher pressures (5–20 atm) were reported in an early investigation of radiative ignition of double-base propellants (DeLuca et al. 1976a). The "approximate" model cannot be used to predict such effects.

These comments emphasize the shortcomings of the "approximate" model and imply that "useful" laser ignition models (for many propellants) need to include explicitly the effect of gas phase reactions on the physics of flamespreading.

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APPENDIX:
DERIVATION

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The derivation of the equations for calculating ignition delays from the "approximate" model (described in reference 4) are reproduced below (with typographical errors corrected).

The energy equations and the initial and boundary conditions for inert heating and adiabatic reaction are:

$$\begin{aligned} \frac{\partial T^I}{\partial t} &= \alpha \frac{\partial^2 T^I}{\partial x^2} + \frac{q_o}{\rho c} n \exp(-nx) \\ T^I(x, 0) &= T_o, \quad T^I(\infty, 0) = T_o, \quad \frac{\partial T^I}{\partial x}(0, t) = 0 \end{aligned} \quad (1a)$$

$$\frac{dT^A}{dt} = \frac{QZ}{c} \exp\left(-\frac{E}{RT^A}\right) \quad T^A(0) = T_1 \quad (1b)$$

where superscripts *I* and *A* refer to inert and adiabatic conditions, respectively. The corresponding nondimensional equations are obtained using the following:

$$\begin{aligned} \theta &= \frac{1}{\beta} \left(\frac{T}{T_a} - 1 \right), \quad \beta = \frac{RT_a}{E}, \quad \xi = \frac{x}{x_a}, \quad \tau = \frac{t}{t_a}, \quad \delta = \frac{q_o}{q_a}, \quad \mu = nx_a \\ x_a &= \sqrt{\alpha x_a}, \quad q_a = \frac{\lambda \beta T_a}{x_a}, \quad t_a = \frac{\beta c T_a}{QZ} \exp(1/\beta), \quad \alpha = \frac{\lambda}{\rho c} \end{aligned}$$

where the subscript *a* refers to standard values. The results are given by 2a and 2b

$$\begin{aligned} \frac{\partial \theta^I}{\partial \tau} &= \frac{\partial^2 \theta^I}{\partial \xi^2} + \delta \mu \exp(-\mu \xi) \\ \theta^I(\xi, 0) &= \theta_o, \quad \theta^I(\infty, t) = \theta_o, \quad \frac{\partial \theta^I}{\partial \xi}(0, t) = 0 \end{aligned} \quad (2a)$$

$$\frac{d\theta^A}{d\tau} = \exp\left(-\frac{\theta^A}{1+\beta\theta^A}\right) \quad \theta^A(0) = \theta_1. \quad (2b)$$

The inert solution is

$$\begin{aligned} \theta'(\xi, \tau) = & \theta_o + 2\delta\sqrt{\tau} \, i\phi^*(y) - \frac{\delta}{\mu} \exp(-\mu\xi) + \\ & \frac{\delta}{2\mu} \exp(-y^2) \left[\exp(w^2)\phi^*(w) + \exp(v^2)\phi^*(v) \right] \end{aligned} \quad (3a)$$

where

$$y = \frac{\xi}{2\sqrt{\tau}}, \quad w = z - y, \quad v = z + y, \quad z = \mu\sqrt{\tau}$$

$$\phi^*(u) = 1 - \phi(u), \quad \phi(u) = \frac{2}{\sqrt{\pi}} \int_0^u e^{-t^2} dt, \quad i\phi^*(u) = \frac{e^{-u^2}}{\sqrt{\pi}} - u\phi^*(u).$$

The adiabatic solution is

$$\tau = \frac{1}{\beta^2 e^{1/\beta}} \left[F(x_1) - F(x) \right] \quad (3b)$$

where

$$\begin{aligned} x_k = & \left\{ \beta \left[1 + \beta\theta^A(\tau_k) \right] \right\}^{-1}, \quad F(u) = Ei(u) - \frac{\exp(u)}{u} \\ Ei(u) = & \int_{-\infty}^u \frac{e^t}{t} dt \end{aligned}$$

for $\xi = 0$

$$\theta'(0, \tau) = \theta_o + \frac{2}{\sqrt{\pi}} \frac{\delta z}{\mu} + \frac{\delta}{\mu} \left[\exp(z^2)\phi^*(z) - 1 \right] \quad (4a)$$

$$\frac{\partial \theta'(0, \tau)}{\partial \xi} = \delta \mu \exp(z^2) \phi^*(z) \quad (5a)$$

The inert heating time (t_h) and the corresponding surface temperature (T_h) are obtained from the "approximate" model assumptions, i.e., for $\xi = 0$ and $\tau = \tau_h$, where $\tau_h = t_h/t_a$

$$\theta'(0, \tau_h) = \theta^A(0) \quad (6a)$$

$$\frac{\partial \theta'}{\partial \tau}(0, \tau_h) = \frac{d\theta^A(0)}{d\tau} \quad (7a)$$

let $T_a = T_h$

$$\theta'(0, \tau_h) = 0 \quad (6b)$$

from (6a) $\theta^A(0) = 0 \quad (6c)$

from (2b) $\frac{d\theta^A(0)}{d\tau} = 1 \quad (2c)$

from (7a) $\frac{\partial \theta'(0, \tau_h)}{\partial \tau} = 1 \quad (7b)$

from (4a) and (6b)

$$-\theta_o = |\theta_o| = \frac{2}{\sqrt{\pi}} \frac{\delta z_h}{\mu} + \frac{\delta}{\mu} \left[\exp(z_h^2) \phi^*(z_h) - 1 \right] \text{ where } z_h = \mu \sqrt{\tau_h} \quad (4b)$$

from (5a) and (7b) $1 = \delta \mu \exp(z_h^2) \phi^*(z_h) \quad (5b)$

$$\text{for } u \gg 1 \quad \exp(u^2) \phi^*(u) \approx \frac{1}{\sqrt{\pi} u} \left(1 - \frac{1}{2u^2} \right) \quad (5c)$$

assume: $z_h \gg 1, \mu^2 |\theta_o| \gg 1$

using (5c), (4b), and (5b)

$$2z_h^2 - \sqrt{\pi} z_h - \mu^2 |\theta_d| = 0 \quad (8)$$

solution of (8) gives

$$\tau_h = \frac{|\theta_d|}{2} + \frac{1}{2\mu} \sqrt{\frac{\pi}{2} |\theta_d|} \quad (9)$$

The equation used to solve for T_h is obtained from (9) and (5b):

$$\delta = \frac{\pi}{4\mu} + \sqrt{\frac{\pi}{2} |\theta_d|} \quad (10)$$

The dimensional form of (10) is quadratic in $\exp(E/RT_h)$. Real values for T_h are given by

$$\exp(E/RT_h) = \frac{\pi}{8} \frac{\lambda \rho Q Z (T_h - T_o)}{q_o^2} \left[1 + \sqrt{1 + \frac{2q_o}{n\lambda(T_h - T_o)}} \right]^2 \quad (11)$$

T_h is obtained by numerical solution of (11).

t_a is calculated using $T_h = T_a$ and t_h is then obtained from (9).

The chemical induction time (t_{ch}) is obtained using the ignition criterion $\frac{d\theta}{d\tau} = \infty$ and (3b) in which is replaced by $\tau - \tau_h$:

$$\tau - \tau_h = \frac{1}{\beta^2 \exp(1/\beta)} [F(x_1) - F(x)] \quad (3c)$$

where

$$x_k = \left\{ \beta [1 + \beta \theta^A (\tau_k - \tau_h)] \right\}^{-1}$$

$$\text{for } u \gg 1 \quad Ei(u) = \frac{\exp(u)}{u^2} \left(1 + \frac{1}{u} + \frac{2}{u^2} \right)$$

then

$$F(u) = \frac{\exp(u)}{u^2} \left(1 + \frac{2}{u} \right)$$

assume

$$\beta \ll 1, \quad x \gg 1, \quad x_1 \gg 1$$

then (3c) can be written as

$$\tau - \tau_h = (1 + 2\beta) \left[\frac{1}{\left(\frac{d\theta^A}{d\tau} \right)_{\tau_1}} - \frac{1}{\left(\frac{d\theta^A}{d\tau} \right)_{\tau}} \right] \quad (3d)$$

for $\tau_1 = \tau_h$

$$\left(\frac{d\theta^A}{d\tau} \right)_{\tau_1} = 1$$

$$\text{for } \tau = \tau_{ig} \quad \left(\frac{d\theta^A}{d\tau} \right)_{\tau} = \infty \quad \text{where } \tau_{ig} = \frac{t_{ig}}{t_a} \text{ and } t_{ig} = \text{ignition delay}$$

from (3d)

$$\tau_{ch} = \tau_{ig} - \tau_h \approx 1 + 2\beta \quad \text{where } \tau_{ch} = \frac{t_{ch}}{t_a}$$

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